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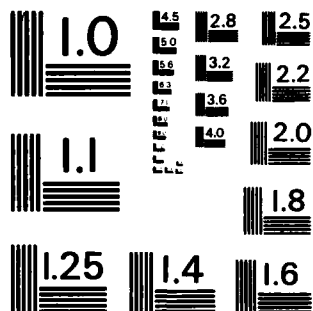
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COMPARISON OF AUTOIONIZATION AND  
PHOTOELECTRON SPECTRA OF CO

By

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) Photoelectron spectra and the autoionization portion of the Auger spectra are compared for CO. Eight final states of the photoelectron spectra coincide with those of the autoionization spectra: three of them are the well-know one-hole states and the other five are two-hole-one-electron states. A photoelectron band at 27.4eV is identified for the first time by its exact match to the strongest band of the autoionization spectra. The symmetries of these and other two-hole-one-electron states are induced by comparing the intensities of X-ray and UV photoelectron spectra. ←		

## 1. Introduction

The two-hole-one-electron (2hle) states of the  $\text{CO}^+$  ion play a significant role in the dissociative photoionization of CO [1]. Photon stimulated desorption of  $\text{CO}^+$  and  $\text{O}^+$  ions from CO chemisorbed on metal surfaces has been found to proceed primarily through these states [2]. These 2hle states have been observed in the autoionization region of the Auger spectra [3,4] as well as in the photoelectron spectra of CO [5,6]. Careful analysis of the two spectra can lead to a better understanding of these 2hle states. In this letter we demonstrate how fruitful a comparison between autoionization spectra (AIS) and photoelectron spectra (PES) can be. The spectra used in this letter are reproduced from published literature. The spectra are scaled to the same energy with comparable intensities.

## 2. Autoionization Spectra

Figs.1a and 1b are portions of the Auger spectra obtained by Ungier and Thomas [3] and Siegbahn et al. [4] respectively. The two spectra are identical, we have included both of them to show that the small peaks and shoulders in each spectrum are indeed reproducible and result from physical processes. The band indicated by a dotted line in Fig.1b arises from the normal Auger transitions, namely the transitions from the  $\text{C}_{1s}$ -hole state to a two-hole state [4,7]. The A bands except the A3 band arise from autoionization initiating from the  $\text{C}_{1s}^{-1}2\pi^*$  state. Ungier and Thomas [3] reached this conclusion by studying the decay of the  $\text{C}_{1s}^{-1}2\pi^*$  state utilizing the (e, 2e) coincidence technique. Moddeman et al. [7] found that all the A bands disappear when <sup>2n</sup>Al K $\alpha$  light source is used and concluded that each  
^

A band originates from a discrete neutral state, including the A3 band. Combining the above two findings, we can conclude that the A3 band originates from the decay of a discrete neutral state other than the  $C_{1s}^{-1}2\pi^*$  state. In Fig.1, we can further confirm these findings by comparing the binding energies of the final states, one obtained from the photoelectron spectrum and the other obtained from the autoionization spectrum assuming that the  $C_{1s}^{-1}2\pi^*$  state is the initial state. Inversely, we can state that the final states of the photoelectron and autoionization spectra are the same for all A bands except the A3 band.

Let us now study the final states of the autoionization process. In autoionization, transitions involving three or more electron jumps are forbidden in first order, since the transition is governed by the two-electron operator,  $1/r_{12}$ . The only accessible final states from the initial state  $C_{1s}^{-1}2\pi^*$  are then the singly ionized states ( $5\sigma^{-1}$ ,  $1\pi^{-1}$ ,  $4\sigma^{-1}$ , and  $3\sigma^{-1}$ ) and the 2hle states with  $le=2\pi^*$ . The singly ionized states can be identified by their binding energies obtained from photoelectron spectra, but none of them have a binding energy in the range 21-32eV. For this reason, we can state that the final states of all the A bands (except the A3 band) have a 2hle electronic configuration with  $le=2\pi^*$ , i.e., the  $2\pi^*$  electron remains as a spectator. This restriction on the final state electron configurations is very useful when attempting to make definite assignments for the A bands which we will do later.

The transition probabilities from the  $C_{1s}^{-1}2\pi^*$  to the  $(2h, 2\pi^*)$  states are approximately the same as those from the  $C_{1s}^{-1}$  state to the 2h states, since the probabilities are identical if the states involved are expressed by a single Slater determinant of identical molecular orbitals. Normal

Auger peaks are more intense than autoionization peaks simply because the  $C_{1s}^{-1}$  state is more populated than the  $C_{1s}^{-1}2\pi^*$  state. In a normal Auger spectra [4,7], three main peaks corresponding to transitions to the  $2h$  states,  $5\sigma^{-1}1\pi^{-1}$ ,  $5\sigma^{-1}4\sigma^{-1}$ , and  $5\sigma^{-2}$ , are observed [8]. In the autoionization spectra, we then expect that the configurations  $5\sigma^{-1}1\pi^{-1}2\pi^*$ ,  $5\sigma^{-1}4\sigma^{-1}2\pi^*$ , and  $5\sigma^{-2}2\pi^*$  give similar intense peaks. Here, one must be aware that the ( $2h$ ,  $2\pi^*$ ) electronic configurations split into more terms than the  $2h$  states. The total intensity of an electronic configuration (e.g.,  $5\sigma^{-1}1\pi^{-1}2\pi^*$ ) will be distributed among the different states (e.g., two  $2\pi^+$  states). Other electronic configurations (e.g.,  $1\pi^{-2}2\pi^*$ ) may gain intensity through configuration interaction (CI) mixing with these three configurations.

The equilibrium position of the  $C_{1s}^{-1}2\pi^*$  state ( $r_e \approx 1.15\text{\AA}$  estimated from NO utilizing the equivalent core approximation) is slightly shifted from the ground state ( $r_e \approx 1.13\text{\AA}$ ). In spite of the small shift, the first vibrational level ( $v=1$ ) of the  $C_{1s}^{-1}2\pi^*$  state gains a significant population via a Frank-Condon transition from the ground state. In their carbon AIS, Moddeman et al. [7] have identified the weak satellite of the  $C_{1s}^{-1}2\pi^* - 1\pi^{-1}$  resonant photoemission band as a "hot" band originating from the vibrationally excited state ( $v=1$ ) of  $C_{1s}^{-1}2\pi^*$ . We identify the weak features just below the A1 and A5 bands (shaded area of Fig.1a and 1b) as hot bands originating from the same vibrational state of  $C_{1s}^{-1}2\pi^*$ . The equilibrium position of the  $C_{1s}^{-1}$  state ( $r_e \approx 1.06\text{\AA}$  estimated from  $NO^+$ ) is far more shifted from the ground state. We would then expect the higher vibrational states of  $C_{1s}^{-1}$  to be populated even more and consequently the hot bands to be more visible. For this reason, we identify the band B (shaded area appearing just before the normal Auger band) as a hot band of the first normal Auger band orig-



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inating from the vibrationally excited states of  $C_{1s}^{-1}$ . This B band is absent in the resonantly excited autoionization spectrum of the  $C_{1s}^{-1}2\pi^*$  state (where the  $C_{1s}^{-1}$  normal Auger spectrum should be absent) [3]; however, such a band is present in the Auger spectrum excited via  $Al\ K\alpha$  light source which does excite the Auger spectrum, but not the AIS spectrum [7]. These two experiments imply that the B band originates from the  $C_{1s}^{-1}$  state supporting our identification.

### 3. Photoelectron Spectra

Fig.1c is a reproduction of the HeII (304Å) photoelectron spectrum obtained by Åsbrink et al. [6]. The original spectrum contained extra lines due to a light source impurity. We have removed those impurity lines according to the original author's interpretation except for the broad structure denoted A5. They have interpreted this band as arising from the N(III) doublet (451.87Å and 452.23Å) impurity lines in the light source. The same band, however, persists in the synchrotron-radiation PES (Fig.1d) which does not have such impurity in its light source. Furthermore, the A5 band appears also in the X-ray PES (Fig.1e) and in the carbon autoionization spectra (Fig.1a and 1b). We therefore, interpret the broad structure of the A5 band in the HeII PES as a real  $CO^+$  band corresponding to a  $2h1e$  state. The shape of this band, i.e., the absence of vibrational structure, suggests that the A5 state is repulsive. The dissociation of CO through this and other  $2h1e$  states has been studied. We will publish the results elsewhere [9].

The first band in the He(II) spectrum, denoted as A1, was contaminated by a light source impurity. Because of this, the presence of the band A1

was not apparent as it now appears in Fig.1c. (Recall that we have removed the impurity lines.) The presence of this band is also not readily apparent in the PES with photon energy 50.3eV (Fig.1d). As a matter of fact, the first (A1) and the second (A2) peaks in Fig.1d have been interpreted [6] as a single band. Here we assert that the second strongest peak (A1) in the carbon autoionization spectrum (Fig.1a and 1b) is supporting evidence for the presence of the A1 band in the photoelectron spectrum.

Table 1 compares the band intensities of three photoelectron spectra excited by different photon energies,  $h\nu=1487$ , 50, and 45eV. These intensities are normalized to the  $3\sigma^{-1}$  intensity for each spectrum. Then the resultant relative band intensities are compared for each band with its own intensity at 1487eV. The numbers in parentheses give that ratio. In other words, we compare the photoelectron spectra keeping the  $3\sigma^{-1}$  intensity constant. Photoelectron spectra using photon energies at 1487eV [10] and 45eV [11] are shown in Fig.2.

In PES, 2hle excitations derive most of their intensities from configuration interaction (CI) mixing with the one-hole (1h) excitations [12,13]. If a 2hle excitation mixes with only one 1h excitation, then their relative intensities should remain the same regardless of the photon energy. The ratio of their intensities is equal to the square of the mixing coefficient. Theoretical calculations [12,13] have shown that the bands at 32eV ( $C_1$  &  $C_2$ ) borrow their intensity mainly from the  $3\sigma^{-1}$  band. However, as seen from Table 1 and Fig.2, the relative intensity of the 32eV band to the  $3\sigma^{-1}$  band is far from constant. In fact, the ratio increases more than twice, when the photon energy is reduced from x-ray (1487eV) to UV (50eV) energies. This increase in ratio indicates that the

bands at 32eV gain their intensity not only from the  $3\sigma^{-1}$  band but also from the  $5\sigma^{-1}$ ,  $1\pi^{-1}$ , and  $4\sigma^{-1}$  bands.

When the photon energy is reduced from 1487eV to 50eV, the  $1\pi^{-1}$  band increases its intensity relative to the other one-hole bands: four times relative to the  $5\sigma^{-1}$  band, five times relative to the  $4\sigma^{-1}$  band, and nineteen times relative to the  $3\sigma^{-1}$  band. The relative intensity gained by the  $1\pi^{-1}$  band is so enormous that we can distinguish the CI mixed  $^2\Pi$  states from the  $^2\Sigma^+$  states. Note that the  $^2\Pi$  states borrow their intensities from the  $1\pi^{-1}$  state and the  $^2\Sigma^+$  states from the  $3\sigma^{-1}$ ,  $4\sigma^{-1}$ , or  $5\sigma^{-1}$  states. Figs.1d and 1e compare the photoelectron spectra having different photon energies, 50.3eV and 1487eV (Al K $\alpha$ ). In Fig.1d, the gained intensity is shown schematically by the shaded areas. We suggest that the shaded intensities originate from  $^2\Pi$  electronic states, and assign the  $^2\Pi$  symmetry to the final states of bands A1 and A5; the  $^2\Sigma^+$  symmetry to those of bands A2, A6, and C1; the  $^2\Pi$  &  $^2\Sigma^+$  to those of the multiple-bands C2. The symmetry of the A4 final state is not obvious. In the 50.3eV PES, it is hard to tell whether the intensity ratio A4 : A2 is larger than that in the Al K $\alpha$  spectra. But the He(II) PES clearly shows that this ratio is larger than that in the Al K $\alpha$  spectra. Therefore we tentatively assign the  $^2\Pi$  symmetry to the final states of the A4 band. Very recently Krummacher et al. [14] have identified the band around 27eV as a  $^2\Pi$  state based on an intensity argument and a theoretical calculation. However, they have not recognized that two overlapping bands are present around 27eV, namely the A5 (27.4eV,  $^2\Pi$ ) and the A6 band (27.9eV,  $^2\Sigma^+$ ).

The band corresponding to the C1 band of PES is missing in the C-AIS. This implies that the electronic configuration of the C1 final state is

not  $(2h, 2\pi^*)$  but  $(2h, 6\sigma^*)$  or  $(2h, \text{Rydberg})$ . A definite assignment of the electronic configurations within the C2 band is difficult for two reasons: first, the presence of the C2 band in the C-AIS is difficult to verify, because all bands in this region are masked by the intense normal Auger bands; second, theoretical calculations indicate that many states are present in this region.

#### 4. Theoretical Calculations.

Theoretical calculations [12,13,15] on the  $^2\Sigma^+$  states of  $\text{CO}^+$  provide consistent results, especially on the lowest five  $^2\Sigma^+$  states, which are identified by the electronic configurations,  $5\sigma^{-1}$ ,  $4\sigma^{-1}$ ,  $5\sigma^{-1}1\pi^{-1}2\pi^*$ ,  $5\sigma^{-1}1\pi^{-1}2\pi^*$ , and  $5\sigma^{-2}6\sigma^*$  in the order of increasing binding energy.

Relying on these theoretical results, we assign electronic configurations to the  $^2\Sigma^+$  states: A2( $5\sigma^{-1}1\pi^{-1}2\pi^*$ ), A6( $5\sigma^{-1}1\pi^{-1}2\pi^*$ ), and C1( $5\sigma^{-2}6\sigma^*$ ). These assignments agree with previous conclusions that A2 and A6 correspond to  $(2h, 2\pi^*)$  electronic configurations and C1 to a  $(2h, \text{Rydberg})$  or  $(2h, 6\sigma^*)$ . The binding energies calculated by the theories also agree very well with the experimental binding energies of the A2, A6, and C1 bands, especially on their relative values.

In contrast with the  $^2\Sigma^+$  states, theoretical calculations [12,16,17] on the  $^2\Pi$  states disagree with each other widely. The predicted binding energy of the second lowest  $^2\Pi$  state (the lowest state being that of  $1\pi^{-1}$ ) differs widely, e.g., 20.3eV (INDO CI[16]), 22.2eV (Full-Valence CI[17]), 22.9eV (Polarization CI[12]), 36.0eV (Green's Function[12]), and 37.2eV (Single-Excitation CI[12]). Not only their predicted binding energies but

also the number of the states predicted to lie between 21 - 35eV is quite different. Furthermore, no electron configurations are available except the  $1\pi^{-2}2\pi^*$  for the  $^2\Pi$  at 20.3eV given in Ref.[16]. In such a circumstances, we can assign only in a general way; a combination of  $5\sigma^{-2}2\pi^*$ ,  $1\pi^{-2}2\pi^*$ , and  $5\sigma^{-1}4\sigma^{-1}2\pi^*$  configurations is assigned to the  $^2\Pi$  bands, A1, A4, and A5.

Table 2 summarizes our conclusions on the 2h1e states of  $\text{CO}^+$  reached by comparing the autoionization and photoelectron spectra of CO.

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### Figure Captions

Figure 1 Comparison between carbon autoionization spectra (C-AIS) and photoelectron spectra (PES) of CO.

- (a) C-AIS by 2.5KeV electron impact (obtained from Ref.[3])
- (b) C-AIS by 5KeV electron impact (obtained from Ref.[4])
- (c) PES with He II ( $h\nu=40.8\text{eV}$ ) (reconstructed from Ref.[6])
- (d) Synchrotron-radiation PES with  $h\nu=50.3\text{eV}$  (obtained from Ref.[5])
- (e) X-ray PES with Al K $\alpha$  ( $h\nu=1487\text{eV}$ ) (obtained from Ref.[10])

Figure 2 Comparison of CO photoelectron spectra.

- (Top) excited by UV photons of 45eV (Ref.[11])
- (Bottom) excited by X-rays (Al K $\alpha$ ) of 1487eV (Ref.[10])

Table 1

Comparison between the relative intensities of photoelectron spectra obtained at photon energies of 1487eV, 50eV, and 45eV.

Band	Binding energy (eV)	Relative intensities		
		$h\nu=1487\text{eV}^{\text{a}}$	$h\nu=50\text{eV}^{\text{b}}$	$h\nu=45\text{eV}^{\text{b}}$
$5\sigma^{-1}$	14.0	22 (1.0) <sup>c</sup>	97 ( 4.4)	129 ( 5.9)
$1\pi^{-1}$	16.9	16 (1.0)	296 (18.5)	360 (22.5)
$4\sigma^{-1}$	19.7	50 (1.0)	181 ( 3.6)	202 ( 4.0)
A1 & A2	$\approx 23$	5 (1.0)	21 ( 4.2)	25 ( 5.0)
C1 & C2	$\approx 32$	25 (1.0)	52 ( 2.1)	61 ( 2.4)
$3\sigma^{-1}$	38.3	100 (1.0)	100 ( 1.0)	100 ( 1.0)

a) Measured from the spectrum of Ref. [10].

b) Data obtained from Ref. [11].

c) Numbers in parentheses give the ratio to the intensity at  $h\nu=1487\text{eV}$ .

Table 2

Initial and final states of bands appearing in the photoelectron and auto-ionization spectra of CO.

Band	Initial state	Final state			Comments
		Binding d) energy(eV)	Symmetry	Electronic configuration	
A1	$C_{1s}^{-1} 2\pi^*$ ( $^1\Pi$ ) for AIS or Ground state for PES	22.6	$^2\Pi$	a)	
A2		23.2	$^2\Sigma^+$	$5\sigma^{-1} 1\pi^{-1} 2\pi^*$	
A4		25.5	$^2\Pi$	a)	b)
A5		27.4	$^2\Pi$	a)	Repulsive
A6		27.9	$^2\Sigma^+$	$5\sigma^{-1} 1\pi^{-1} 2\pi^*$	c)
A3	Discrete neutral state	? e)	?	?	Absent in PES
B	$C_{1s}^{-1}$ ( $v \geq 1$ )	41.9 f)	$^1\Pi$	$5\sigma^{-1} 1\pi^{-1}$	Hot band
C1	ground state	$\approx 30$	$^2\Sigma^+$	$5\sigma^{-2} 6\sigma^*$	Absent in AIS
C2	ground state	$\approx 32$	$^2\Pi$ & $^2\Sigma^+$	?	Multiple band

a) A combination of  $5\sigma^{-2} 2\pi^*$ ,  $5\sigma^{-1} 4\sigma^{-1} 2\pi^*$  and  $1\pi^{-2} 2\pi^*$  states.

b) Vibrational progression observed in He II PES is similar to that of A1.

c) Vibrational progression observed in He II PES is similar to that of A2.

d) Vertical binding energies.

e) This binding energy cannot be calculated, because the initial state energy is not known.

f) This binding energy is different from that indicated in Fig.1, because the Fig.1a and 1b binding energy scale is appropriate only for the  $C_{1s}^{-1} 2\pi^*$  initial state.

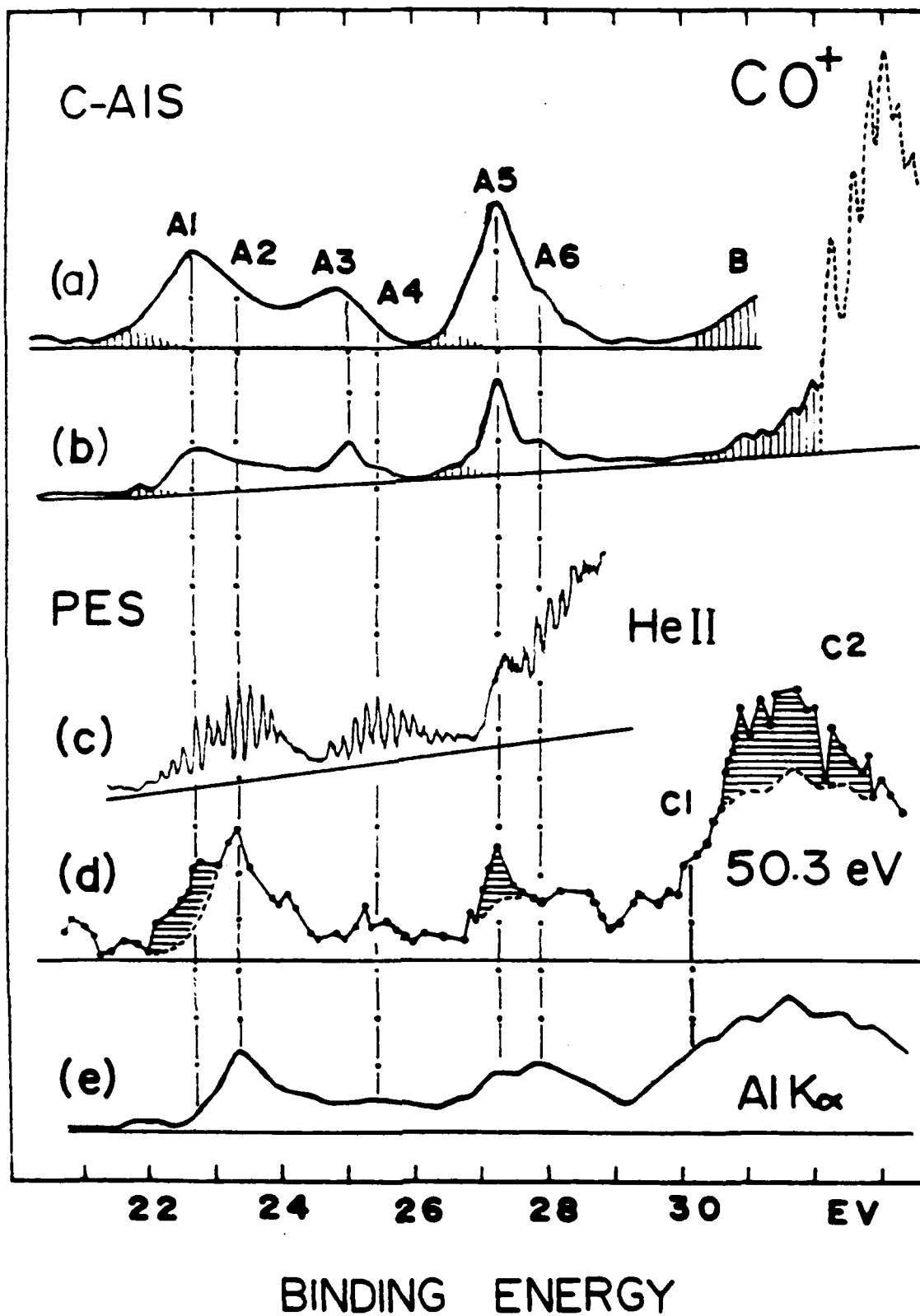


Figure 1

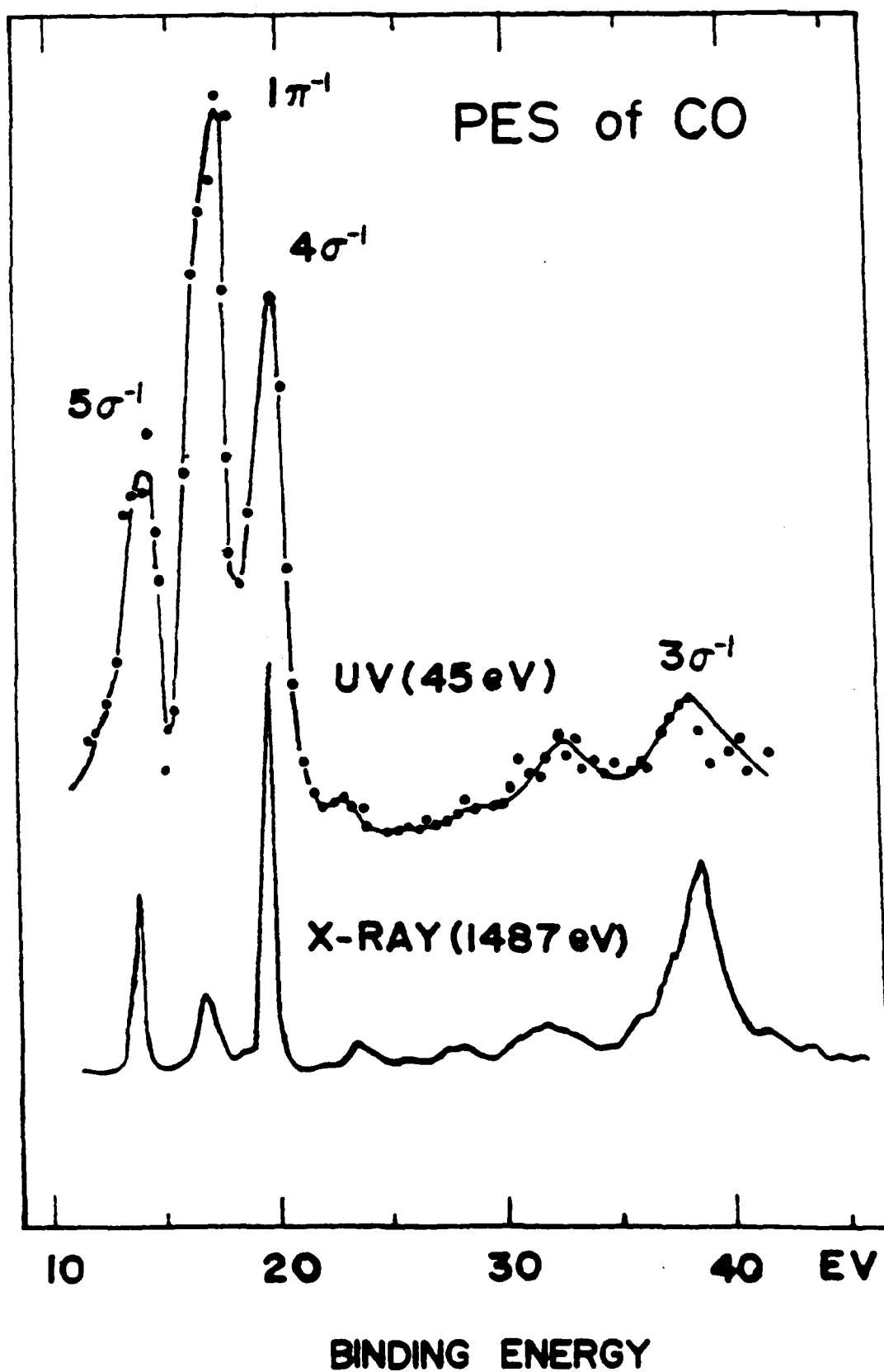


Figure 2

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